

CONFIRMATION OF CIRCUMSTELLAR PHOSPHINE

M. AGÚNDEZ¹, J. CERNICARO¹, L. DECIN^{2,3}, P. ENCRENAZ⁴, AND D. TEYSSIER⁵

¹ Instituto de Ciencia de Materiales de Madrid, CSIC, C/ Sor Juana Inés de la Cruz 3, E-28049 Cantoblanco, Spain

² Sterrenkundig Instituut Anton Pannekoek, University of Amsterdam, Science Park 904, NL-1098 Amsterdam, The Netherlands

³ Instituut voor Sterrenkunde, Katholieke Universiteit Leuven, Celestijnenlaan 200D, B-3001 Leuven, Belgium

⁴ LERMA, Observatoire de Paris, 61 Av. de l'Observatoire, F-75014 Paris, France

⁵ European Space Astronomy Centre, Urb. Villafranca del Castillo, P.O. Box 50727, E-28080 Madrid, Spain

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ABSTRACT

Phosphine (PH₃) was tentatively identified a few years ago in the carbon star envelopes IRC +10216 and CRL 2688 from observations of an emission line at 266.9 GHz attributable to the $J = 1-0$ rotational transition. We report the detection of the $J = 2-1$ rotational transition of PH₃ in IRC +10216 using the HIFI instrument on board *Herschel*, which definitively confirms the identification of PH₃. Radiative transfer calculations indicate that infrared pumping in excited vibrational states plays an important role in the excitation of PH₃ in the envelope of IRC +10216, and that the observed lines are consistent with phosphine being formed anywhere between the star and 100 R_* from the star, with an abundance of 10^{-8} relative to H₂. The detection of PH₃ challenges chemical models, none of which offer a satisfactory formation scenario. Although PH₃ holds just 2% of the total available phosphorus in IRC +10216, it is, together with HCP, one of the major gas phase carriers of phosphorus in the inner circumstellar layers, suggesting that it could also be an important phosphorus species in other astronomical environments. This is the first unambiguous detection of PH₃ outside the solar system, and is a further step toward a better understanding of the chemistry of phosphorus in space.

Key words: astrochemistry – circumstellar matter – line: identification – molecular processes – stars: AGB and post-AGB – stars: individual (IRC +10216)

Online-only material: color figure

1. INTRODUCTION

Phosphorus is a biogenic element, so studying the various forms in which it is present in different astronomical environments is a matter of great interest, although still poorly understood (Maciá 2005). Only five P-containing molecules (PN, CP, HCP, PO, and C₂P) have been observed in the gas phase of interstellar and circumstellar media (Turner & Bally 1987; Ziurys 1987; Guélin et al. 1990; Agúndez et al. 2007; Tenenbaum et al. 2007; Halfen et al. 2008). These five molecules are all observed in circumstellar envelopes around evolved stars. However, in interstellar clouds only PN is observed and it has a relatively low abundance, implying that most of the phosphorus must be elsewhere, probably depleted in dust grains (Turner et al. 1990).

Phosphine (PH₃), the phosphorus cousin of ammonia, is a relatively stable molecule that could be holding an important fraction of the phosphorus in various astronomical environments. Since more than 30 yr ago, PH₃ has been known to be present in the atmospheres of the giant gaseous planets Jupiter and Saturn, where it is the major phosphorus carrier (Bregman et al. 1975; Larson et al. 1977; Weisstein & Serabyn 1996). Upper limits obtained in Neptune and Uranus imply that the gas phase abundance of phosphorus is probably subsolar in the atmospheres of these icy giants (Moreno et al. 2009). Phosphine ice is also a plausible major phosphorus constituent of comets, although recent searches in the gaseous coma of a few comets were unsuccessful, providing upper limits that were not significant enough to conclude whether or not PH₃ holds most of phosphorus in these solar system bodies (Crovisier et al. 2004; Agúndez et al. 2014).

Outside the solar system, no evidence has been found of the presence of PH₃ in interstellar clouds, either in the gas phase or as ice in dust grains. There is, however, some evidence

of PH₃ being present in the outflows of evolved stars. A few years ago this molecule was tentatively identified in the carbon star envelope IRC +10216. The identification was based on the $J = 1-0$ rotational transition, lying at 266.9 GHz, which, however, appeared contaminated by a narrow line assigned to SiS in its $v = 4$ vibrational state (Agúndez et al. 2008). An emission feature attributable to the $J = 1-0$ line of PH₃ was also observed independently by Tenenbaum & Ziurys (2008) in the spectra of IRC +10216 and the carbon-rich envelope CRL 2688. The observation of just one line, affected by blending in the case of IRC +10216, led them to consider the detection of PH₃ as tentative. Here, we report the detection of the $J = 2-1$ rotational transition of PH₃ in IRC +10216 using the HIFI instrument on board *Herschel*,⁶ which definitively confirms the identification of phosphine in this source. This is the first time that PH₃ is unambiguously observed outside the solar system.

2. OBSERVATIONS: IDENTIFICATION OF PH₃

The HIFI observations of IRC +10216 were obtained in 2010 May and from October to December, in the context of two GT1 programs dedicated to performing a line survey in all HIFI bands and searching for light hydrides at selected frequencies. Data were taken in double beam-switching mode with a spectral resolution of 1.1 MHz and a channel spacing of 0.5 MHz, and processed using the standard *Herschel* pipeline up to level 2, which provides fully calibrated spectra in the antenna temperature scale. The intensity scale was later transformed to the main beam brightness temperature T_{MB} . The local oscillator was shifted in frequency to identify any emission arising from

⁶ *Herschel* is an ESA space observatory with science instruments provided by European-led Principal Investigator consortia and with important participation from NASA.

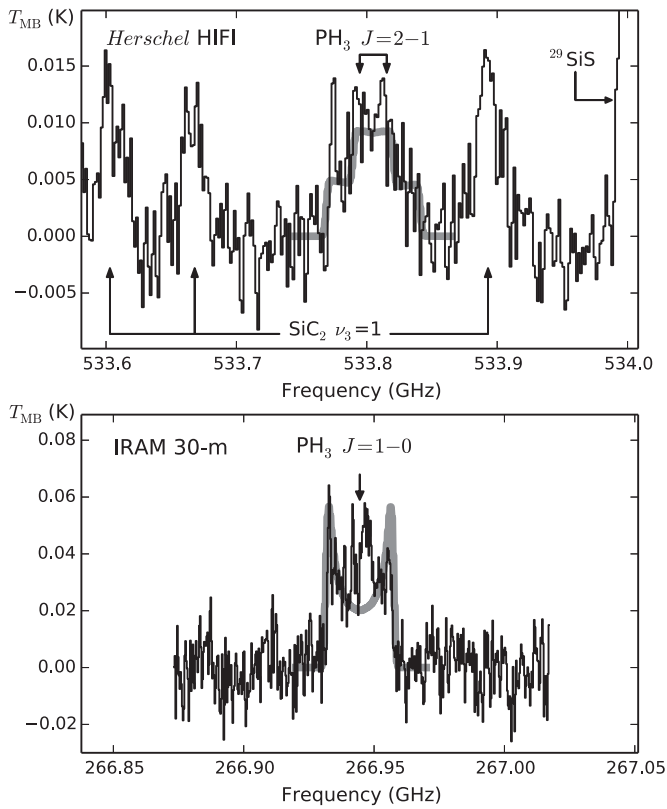


Figure 1. PH_3 lines observed in IRC +10216 (black histogram). The lower panel shows the $J = 1-0$ transition at 266.9 GHz observed with the IRAM 30 m telescope at a spectral resolution of 0.32 MHz. An overlapping narrow line arising from $\text{SiS } v = 4$ has been subtracted (see Agúndez et al. 2008). The upper panel shows the $J = 2-1$ transition (consisting of $K = 0$ and $K = 1$ components) at 533.8 GHz observed with *Herschel* HIFI. The spectrum, smoothed to a spectral resolution of 1.5 MHz, also shows three lines of $\text{SiC}_2 v_3 = 1$ and one line of ^{29}SiS . Thick gray lines are the line profiles calculated with the LVG model (see Section 3). The central peak observed in the $J = 1-0$ line, not accounted for by the model, is probably due to a non-optimal subtraction of the $\text{SiS } v = 4$ overlapping line.

the image band. Spectra were smoothed to a spectral resolution of 1.5 MHz. For details about the data reduction, we refer to Cernicharo et al. (2010).

Phosphine was searched for in IRC +10216 with HIFI through its $J = 2-1$ and $J = 3-2$ rotational transitions. This molecule is an oblate symmetric rotor and thus its rotational levels are defined by the quantum numbers J and K , and radiative transitions are only allowed within levels of the same K ladder. The K ladders are grouped into two distinct forms: ortho and para, between which radiative and collisional transitions are severely forbidden. The rotational spectrum of PH_3 has been extensively studied in the laboratory and line frequencies are accurately known (Cazzoli & Puzzarini 2006; Müller 2013; Sousa-Silva et al. 2013). Its electric dipole moment has been measured as 0.574 D (Davies et al. 1971).

The $J = 2-1$ rotational transition of PH_3 consists of two K components: 2_0-1_0 at 533.7946 GHz and 2_1-1_1 at 533.8153 GHz. The HIFI spectrum of IRC +10216 at these frequencies is shown in the upper panel of Figure 1. The T_{MB} rms noise level is 0.0024 K per 1.5 MHz channel. There is an emission feature with $T_{\text{MB}} \sim 0.01$ K centered at 533.8 GHz, which can be clearly identified with a composite of the two K components of the $J = 2-1$ transition of PH_3 . We have verified that there is no other obvious assignment for this emission feature by looking at the spectral catalog of

MADEX (Cernicharo 2012). Each of the two K components has a line width consistent with an expansion velocity v_{exp} of 14.5 km s $^{-1}$, as do most of the lines arising from the outer envelope in IRC +10216 (Cernicharo et al. 2000), including the line previously observed with the IRAM 30 m telescope and assigned to the $J = 1-0$ transition of PH_3 (see the lower panel of Figure 1 and Agúndez et al. 2008). The spectrum around 533.8 GHz also shows three narrow lines, with $v_{\text{exp}} \sim 8-10$ km s $^{-1}$, arising from SiC_2 in its $v_3 = 1$ vibrational state and one from ^{29}SiS . Lines from these species have been observed already in IRC +10216 at millimeter wavelengths (Cernicharo et al. 2000; Agúndez et al. 2012) and are also detected across the HIFI spectral scan.

The $J = 3-2$ transition of PH_3 lying at 800.5 GHz is not detected because the sensitivity of the HIFI spectrum at these frequencies is not good enough. The T_{MB} rms noise level is 20 mK per 1.5 MHz channel and this PH_3 line is expected to have a T_{MB} of some mK. Moreover, even if the sensitivity were much better, the detection of this line would be hampered by the fact that the $K = 0$ and $K = 1$ components, at 800.4562 GHz and 800.4871 GHz, would appear severely blended with two strong lines from SiC_2 ($34_{10,25}-33_{10,24}$ and $34_{10,24}-33_{10,23}$ at 800.4842 GHz and 800.4907 GHz; Müller et al. 2012). Intense lines from warm SiC_2 are numerous across the whole HIFI spectrum (Cernicharo et al. 2010). The $K = 2$ component of PH_3 at 800.5799 GHz does not overlap with these SiC_2 lines (the line widths in IRC +10216's spectra at these frequencies are ~ 80 MHz) and thus would be observable if the sensitivity were good enough.

The detection of the $J = 2-1$ transition of PH_3 with HIFI, together with the previous detection of the $J = 1-0$ line (Agúndez et al. 2008; Tenenbaum & Ziurys 2008), definitively confirms the identification of PH_3 in IRC +10216. The nondetection of the $J = 3-2$ line in our HIFI data is consistent with the line intensities of the $J = 1-0$ and $J = 2-1$ lines, and thus with the identification of PH_3 .

3. DISCUSSION

To extract information about the abundance, spatial distribution, and excitation of PH_3 in the envelope of IRC +10216, the observed lines must be compared with the results of excitation and radiative transfer calculations. Here, the calculations are based on a multi-shell large velocity gradient (LVG) formalism and the model of the envelope adopted is that of Agúndez et al. (2012). Basically, it consists of an asymptotic giant branch (AGB) star surrounded by a spherically expanding envelope of gas and dust. The adopted distance and mass-loss rate are 130 pc and $2 \times 10^{-5} M_{\odot} \text{ yr}^{-1}$, and dust is assumed to condense at a radius of $5 R_{*}$. The stellar properties, radial profiles, and dust parameters are described in Agúndez et al. (2012). In the innermost layers ($< 5 R_{*}$), we have adopted the downward revision of the particle density derived by Cernicharo et al. (2013) from ALMA observations of HNC in various excited vibrational states.

Energy levels and transition frequencies of PH_3 in its ground vibrational state were computed from the rotational constants reported by Cazzoli & Puzzarini (2006). Line strengths for pure rotational transitions were computed from the dipole moment measured by Davies et al. (1971). Both ortho and para forms of PH_3 were considered with the statistical ortho-to-para ratio of 1. Since rate coefficients for collisional excitation of PH_3 with H_2 or He are not known, we adopted those computed for NH_3 (Danby et al. 1988; Machin & Roueff 2005), properly

corrected to a case without inversion doubling. Scaling of the rate coefficients due to the mass change from NH_3 to PH_3 is very small ($<5\%$) and was thus not considered. No extrapolation was performed for the temperature, and thus the rate coefficients at 300 K, the highest temperature available in the quantum calculations of NH_3 , were adopted at higher temperatures. The use of these rate coefficients is probably a major source of the uncertainty introduced in the excitation calculations. We included rotational levels up to $J_K = 7_6$ for the ortho species and $J_K = 5_5$ for para PH_3 , the highest levels involved in quantum calculations of the collisional rate coefficients for NH_3 . Infrared pumping in excited vibrational states was taken into account by including the first excited states of the four vibrational modes ν_1 , ν_2 , ν_3 , and ν_4 , lying at 2321, 992, 2327, and 1118 cm^{-1} , respectively, over the ground state. Spectroscopic constants and band intensities were taken from laboratory measurements (Baldacci et al. 1980; Tarrago et al. 1984; Fusina & Di Lonardo 2000; Brown et al. 2002; Yurchenko et al. 2006; Sousa-Silva et al. 2013). The fundamental bands of these modes lie at 4.3, 9, and 10 μm , wavelengths at which the flux in the envelope of IRC +10216 is large (Cernicharo et al. 1999). Collisional rates for rovibrational transitions were assumed to be negligible compared with radiative rates.

The spatial distribution of phosphine in the envelope of IRC +10216 is uncertain because the observed line profiles provide limited information. In the case of the $J = 1-0$ line, subtraction of the blended line $\text{SiS } v = 4$ introduces an important uncertainty in the real shape of the PH_3 contribution. The profile of the $J = 2-1$ line is complicated by the overlap of the $K = 0$ and $K = 1$ components and the limited signal-to-noise ratio. In any case, the PH_3 lines observed show a width consistent with an expansion velocity of 14.5 km s^{-1} , which indicates that an important part of the PH_3 emission comes from regions where the gas has already reached the terminal expansion velocity of the envelope. Whether phosphine is formed close to the star or farther out in the envelope is unknown, although there are reasons to suspect that it is not formed too close to the star. On one hand, the abundance of PH_3 predicted at thermochemical equilibrium in the hot and dense surroundings of the star is negligible, $<10^{-12}$ relative to H_2 (Agúndez et al. 2007). On the other, infrared observations rule out the presence of an important amount of the related molecule NH_3 inside a radius of $\sim 20 R_*$ (Keady & Ridgway 1993; Monnier et al. 2000). Following Hasegawa et al. (2006) and Agúndez et al. (2008), here we assume that PH_3 is formed at an inner radius of $20 R_*$. The outer boundary is calculated with a photochemical model in which PH_3 is photodissociated by the external UV field using the rate guessed by MacKay & Charnley (2001). We find that to reproduce the observed lines (see Figure 1), we need a PH_3 abundance of 10^{-8} relative to H_2 , which is slightly higher than the value previously derived by Agúndez et al. (2008) from the $J = 1-0$ line. The adopted abundance profile is shown in the upper panel of Figure 2.

Various conclusions can be drawn from a careful inspection of the excitation and radiative transfer calculations. The first is that infrared pumping in excited vibrational states plays an important role in the excitation of PH_3 in the envelope. It increases the excitation temperature of rotational transitions within the ground vibrational state, and therefore their line intensities, especially the $J = 2-1$ and higher J transitions, and extends the emitting region of the lines to larger radii. This can be appreciated in the lower panel of Figure 2, where we show the contribution to the velocity-integrated line intensity of rays arising at each

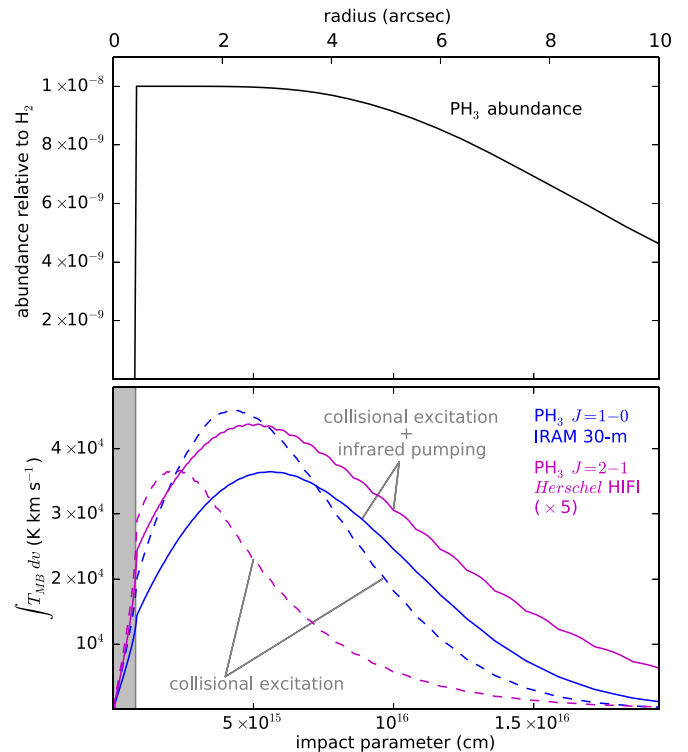


Figure 2. Upper panel: fractional abundance radial profile adopted for PH_3 . Phosphine is assumed to be present from $20 R_*$ with an abundance of 10^{-8} relative to H_2 and to decrease in the outer layers due to photodissociation by interstellar UV photons. Lower panel: velocity-integrated intensity computed as a function of the impact parameter for the $J = 1-0$ and $J = 2-1$ lines of PH_3 . The integration over the impact parameter (if expressed as angular distance in radians) of the magnitude plotted in the ordinate axis yields a calculated value of $\int T_{\text{MB}} dv$. The shaded area corresponds to the region inside $20 R_*$. The two panels share a common abscissa axis.

(A color version of this figure is available in the online journal.)

impact parameter. It can be seen that the bulk of the emission in the $J = 1-0$ and $J = 2-1$ lines comes from regions with impact parameters from the star between $1''$ and $6''$. That is, the emission size of the $J = 1-0$ line is of the order of the main beam of the IRAM 30 m telescope ($9''.2$ at 266.9 GHz), while the $J = 2-1$ line is highly diluted in the main beam of HIFI ($40''$ at 533.8 GHz). We can also appreciate that the observed lines of PH_3 do not trace the regions inside $20 R_*$. Even if we extended phosphine down to the stellar photosphere, the contribution of impact parameters closer than $20 R_*$ to the velocity-integrated line intensities would be less than 5%. In fact, our LVG calculations indicate that the observed lines are compatible with PH_3 being formed anywhere between the stellar surface and $\sim 100 R_*$.

Assuming that the elemental abundance of phosphorus in IRC +10216 is solar ($\text{P}/\text{H} = 2.6 \times 10^{-7}$; Asplund et al. 2009), it turns out that HCP holds 5% of the phosphorus (Agúndez et al. 2007, 2012) while PH_3 holds 2%. It is likely that the rest of the P is in some condensed form as a part of dust grains. In envelopes around other evolved stars, gas phase molecules seem to hold a greater fraction of phosphorus than in IRC +10216. Around half of the P is in the form of HCP and PH_3 in the carbon-rich envelope CRL 2688 (Milam et al. 2008; Tenenbaum & Ziurys 2008), while in oxygen-rich objects PO and PN contain a good fraction of the available P, around 1/4 in VY CMa (Tenenbaum et al. 2007; Milam et al. 2008) and essentially all available phosphorus in IK Tau (De Beck et al. 2013).

In IRC +10216, phosphine is formed somewhere between 1 and 100 R_* from the star, although it is not clear which is the main formation mechanism. Thermochemical equilibrium calculations indicate that its abundance in the surroundings of the star is very low ($<10^{-12}$ relative to H_2) and gas phase chemical kinetics models yield no net formation in the outer layers of the envelope (Agúndez et al. 2007). There are various plausible non-equilibrium processes at work in the inner envelope, which may be at the origin of PH_3 and other hydrides such as NH_3 and H_2O , for which thermochemical equilibrium predicts abundances much lower than observed. Interestingly, the NH_3/PH_3 abundance ratio in IRC +10216 (~ 200 ; Hasegawa et al. 2006) is similar to the solar elemental N/P abundance ratio, indicating that a similar fraction of N and P are locked in NH_3 and PH_3 , respectively. A non-equilibrium chemistry driven by shocks induced by the stellar pulsation has been proposed as a source of water vapor in IRC +10216 (Cherchneff 2011). In this scenario, however, NH_3 is formed with a very low abundance and details on whether or not PH_3 is formed are not provided. Another mechanism proposed for the formation of hydrides such as H_2O and NH_3 in the inner layers of carbon-rich envelopes such as IRC +10216 is photochemistry driven by the penetration of interstellar UV photons across the clumpy envelope (Decin et al. 2010; Agúndez et al. 2010). In this scenario, PH_3 is not efficiently formed, although it is likely due to the lack of relevant chemical kinetics data for phosphorus species. Another possible source of hydrides such as PH_3 that is yet to be explored could be through chemical reactions taking place on dust grain surfaces. Which of these mechanisms, if any, is responsible for the formation of PH_3 in the ejecta of evolved stars such as IRC +10216 has yet to be investigated.

4. SUMMARY

We use the HIFI instrument on board *Herschel* to observe the $J = 2-1$ line of PH_3 in IRC +10216, which, together with the previous observation of the $J = 1-0$ line with the IRAM 30 m telescope, definitively confirms the first identification of PH_3 outside the solar system. Excitation and radiative transfer calculations indicate that the observed lines are consistent with phosphine being formed in the circumstellar envelope anywhere between 1 and 100 R_* from the star with an abundance of 10^{-8} relative to H_2 . The detection of PH_3 challenges chemical models, as no obvious formation route has for the moment been found. Prospects to put further constraints on the distribution and origin of PH_3 in IRC +10216 with telescope facilities such as ALMA seem challenging. The $J = 1-0$ line is blended with a SiS $v = 4$ line, which makes it complicated to disentangle the contribution from each line to the spatial distribution of the emission. The $J = 2-1$ line at 533.8 GHz cannot be reached from the ground because of the terrestrial atmospheric opacity, and the $J = 3-2$ line is also severely blended with a strong line of SiC_2 . Although PH_3 holds a minor fraction of phosphorus in IRC +10216, it is, together with HCP, one of the major gas phase carriers of phosphorus in the inner circumstellar layers, suggesting that it could also be an important phosphorus species in other astronomical environments. Observations of PH_3 in sources other than IRC +10216, such as CRL 2688, may help us to better understand its formation and the implications for the chemistry of phosphorus in space.

HIFI has been designed and built by a consortium of institutes and university departments from across Europe, Canada, and the United States (NASA) under the leadership of SRON, Netherlands Institute for Space Research, Groningen, The Netherlands, and with major contributions from Germany, France, and the U.S. Consortium members are Canada: CSA, U. Waterloo; France: CESR, LAB, LERMA, IRAM; Germany: KOSMA, MPIfR, MPS; Ireland: NUI Maynooth; Italy: ASI, IFSI-INAF, Osservatorio Astrofisico di Arcetri-INAF; Netherlands: SRON, TUD; Poland: CAMK, CBK; Spain: Observatorio Astronómico Nacional (IGN), Centro de Astrobiología (INTA-CSIC); Sweden: Chalmers University of Technology - MC2, RSS, & GARD, Onsala Space Observatory, Swedish National Space Board, Stockholm University-Stockholm Observatory; Switzerland: ETH Zurich, FHNW; USA: CalTech, JPL, NHSC. M.A. and J.C. thank Spanish MICINN for funding support through grant CSD2009-00038.

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